DECLARATION

I, Ryuichi YAMADA, a Japanese Patent Attorney registered No. 7898 having my Business Office at Hasegawa Bldg., 4F, 7-7 Toranomon 3-chome, Minato-ku, Tokyo, Japan, solemnly and sincerely declare:

That I have a thorough knowledge of Japanese and English languages; and

That the attached pages contain a correct translation into English of the specification of the following Japanese Application:

APPLICATION NUMBER	DATE OF APPLICATION
190866/2001(Pat.)	25/JUN/2001

Applicant(s)

CANON KABUSHIKI KAISHA

Signed this 16th day of September, 2003.

Ryuichi YAMADA

PATENT OFFICE

JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following application as filed with this Office.

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190866/2001(Pat.)	25/JUN/2001

Applicant(s)

CANON KABUSHIKI KAISHA

19/OCT/2001

Director-General,
Patent Office : KOZO OIKAWA (Seal)

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[Document]

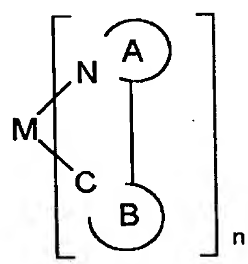
Specification

[Title of the Invention]

Luminescence Device and Display Apparatus

[Claims]

1. A luminescence device, comprising: an organic compound layer comprising a metal coordination compound represented by the following formula (1):



{wherein M denotes Ir, Rh or Pd; n is 2 or 3; A and B are a nitrogen atom and a carbon atom, respectively, connected to a metal atom M; A and B are cyclic groups containing a nitrogen atom and a carbon atom, respectively, and capable of having a substituent, A and B being connected to each other via a covalent bond, [said substituent being a halogen atom; nitro group; a trialkylsilyl group (of which the alkyl group is a linear or branched alkyl group having 1 - 8 carbon atoms); or a linear or branched alkyl group having 1 - 20 carbon atoms (of which the alkyl group includes one or at least two non-neighboring methylene

groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and includes a hydrogen atom which can be replaced with a fluorine atom] with the proviso that a sum of nitrogen atom and sulfur atom present in ring structures of A and B is at least 2}..

2. A device according to Claim 1, wherein the metal coordination compound of formula (1) has a molecular structure free from a portion substantially causing intramolecular rotation and exhibits a peak emission wavelength of at least 550 nm.

3. A device according to Claim 1 or 2, wherein the metal coordination compound of formula (1) contains a ligand having a dipole moment of at most 7 debye and exhibits a peak emission wavelength of at least 550 nm.

4. A device according to Claim 3, wherein the ligand has a dipole moment of at most 4 debye.

5. A luminescence device, comprising an organic compound layer comprising a metal coordination compound, wherein the metal coordination compound has a molecular structure free from a portion substantially causing intramolecular rotation and

exhibits a peak emission wavelength of at least 550 nm.

6. A luminescence device, comprising an organic compound layer comprising a metal coordination compound, wherein the metal coordination compound contains a ligand having a dipole moment of at most 7 debye and exhibits a peak emission wavelength of at least 550 nm.

7. A device according to Claim 6, wherein the ligand has a dipole moment of at most 4 debye.

8. A device according to any one of Claims 1 - 7, further comprising two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the pair of electrodes to produce luminescence.

9. A display apparatus, comprising: a luminescence device according to any one of Claims 1 - 8 and a portion for driving the luminescence device.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Pertains]

The present invention relates to a luminescence device using the compound. More specifically, the present invention relates to a luminescence device employing a metal coordination compound as a luminescence material so as to allow stable and high luminescence efficiency.

[0002]

[Prior Art]

An applied research of an organic electroluminescence (EL) device has been energestically studied as a luminescence device with a high responsiveness and high efficiency.

Basic structures thereof are shown in Figures 1(a) and 1(b) (e.g., as described in "Macromol. Symp.", 125, pp. 1 - 48 (1997)).

[0003]

As shown in Figure 1, the organic EL device generally has a structure including a transparent electrode 14, a metal electrode 11, and a plurality of organic film layers disposed on a transparent substrate 15.

[0004]

Referring to Figure 1(a), the organic layers include a luminescence layer 12 and a hole transport layer 13. The transparent electrode 14 may be formed of a film of ITO (indium tin oxide) having a larger work function to ensure a good hole injection

performance from the transparent electrode 14 into the hole transport layer 13. On the other hand, the metal electrode 11 may be formed of a layer of aluminum, magnesium, alloys thereof, etc., having a smaller work function to ensure a good electron injection performance into the organic layer(s). These electrodes may be formed in a thickness of 50 - 200 nm.

[0005]

The luminescence layer 12 may be formed of, e.g., aluminum quinolinol complex (representative example thereof may include Alq3 shown below having an electron transporting characteristic and a luminescent characteristic. The hole transport layer 13 may be formed of, e.g., triphenyldiamine derivative (representative example thereof may include α-NPD shown below having an electron donating characteristic.

[0006]

The above-structured device exhibits a rectification characteristic, so that when an electric field is applied between the metal electrode 11 as a cathode and the transparent electrode 14 as an anode, electrons are injected from the metal electrode 11 into the luminescence layer 12 and holes are injected from the transparent electrodes 14.

[0007]

The injected holes and electrons are recombined within the luminescence layer 12 to form excitons, thus producing luminescence. At this time, the hole transport layer 13 functions as an electron-blocking layer to increase a recombination efficiency at the boundary between the luminescence layer 12 and the hole transport layer 13, thus enhancing a luminescence efficiency.

[0008]

Referring to Figure 1(b), an electron transport layer 16 is disposed between the metal electrode 11 and the luminescence layer 12, whereby an effective carrier blocking performance can be ensured by separating functions of luminescence, electron transport and hole transport, thus allowing efficient luminescence. The electron transport layer 16 may be formed of, e.g., oxadiazole derivatives.

[0009]

In ordinary organic EL devices, heretofore, fluorescence produced during a transition of luminescent center molecule from a singlet excited state to a ground state is used as luminescence. On the other hand, not the above fluorescence (luminescence) via singlet exciton, phosphorescence (luminescence) via triplet exciton has been studied for use in organic EL device. Representative published literature may include Article 1: "Improved

energy transfer in electrophosphorescent device" (D.F. O'Brien et al., Applied Physics Letters, Vol. 74, No. 3, pp. 442 - (1999)) and Article 2: "Very high-efficiency green organic light-emitting devices based on electrophosphorescence" (M.A. Baldo et al., Applied Physics Letters, Vol. 75, No. 1, pp. 4 - (1999)).

The EL devices shown in these documents may generally have a four-layer structure shown in Figure 1(c), which structure includes a hole transfer layer 13, a luminescence layer 12, an exciton diffusion-prevention layer 17, and an electron transport layer 16. Materials used therein are carrier transfer materials and phosphorescent materials shown below. Abbreviations of the respective materials are shown below.

Alq3: aluminum-quinolinol complex,

α-NPD: N4,N4'-di-naphthalene-1-yl-N4,N4'-diphenyl-biphenyl-4,4'-diamine,

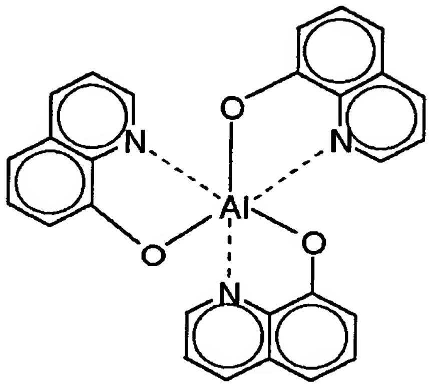
CBP: 4,4'-N,N'-dicarbazole-biphenyl,

BCP: 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline,

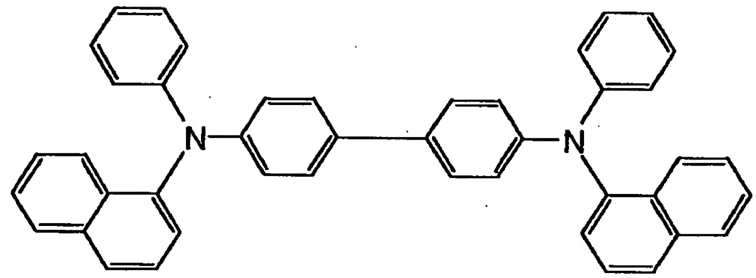
PtEOP: platinum-octaethyl porphine complex, and

$Ir(ppy)_3$: iridium-phenylpyridine complex.

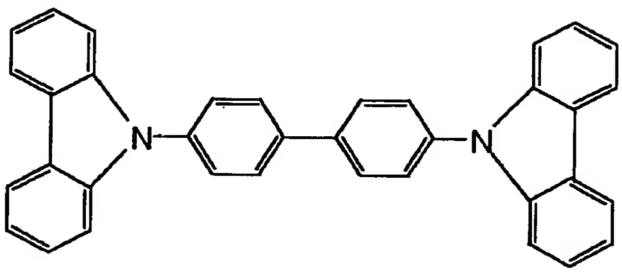
[0011]



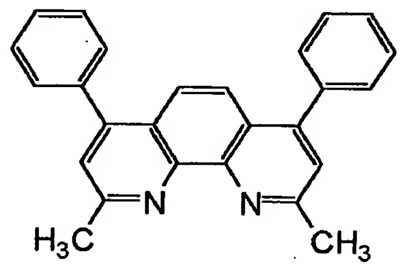
Alq3



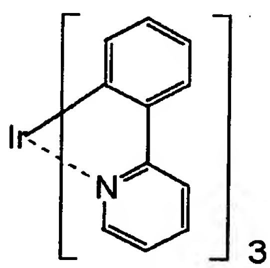
α -NPD



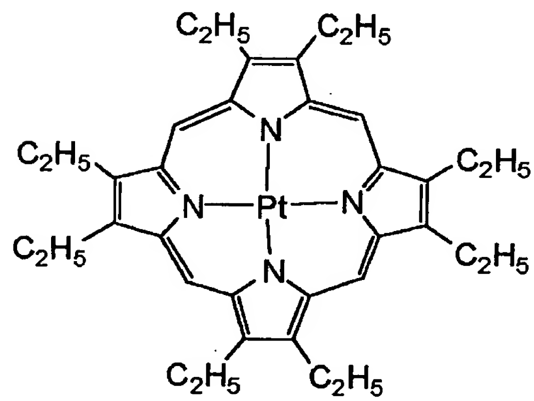
CBP



BCP



$Ir(ppy)_3$



PtOEP

[0012]

In both the Articles 1 and 2, higher efficiencies have been achieved by using a layer structure including a hole transport layer 13 of α-NPD, an electron transport layer 16 of Alq3, an exciton diffusion-prevention layer 17 of BPC, and a luminescence layer 12 of a mixture of CPB as a host material with $Ir(ppy)_3$ or PtOEP as a (guest) phosphorescence material doped into CBP at a concentration of ca. 6 wt. %.

[0013]

The reason why the phosphorescence (luminescence) material has particularly attracted notice is that the phosphorescence material is expected to provide a higher luminescence efficiency in principle. This is because excitons produced by recombination of carriers comprise singlet excitons and triplet excitons presented in a ratio of 1:3. In the conventional organic EL device, fluorescence at the time of transition from the singlet excited state to the ground state is utilized as luminescence. A resultant luminescence efficiency is 25 % (which is an upper limit) based on the number of the produced excitons in principle. However, if phosphorescence from excitons generated from the triplet excited state is used, a resultant luminescence efficiency is expected to be at least three times that of the case

of fluorescence in principle. In addition thereto, if intersystem crossing from the singlet excited state which is a higher energy level to the triplet excited state is taken into consideration, the luminescence efficiency of phosphorescence can be expected to be 100 % (four times that of fluorescence) in principle.

[0014]

Other documents describing phosphorescence from the triplet excited state may include, e.g., Japanese Laid-Open Patent Application (JP-A) 11-329739 (organic EL device and production process thereof), JP-A 11-256148 (luminescent device and organic EL device) and JP-A 8-319482 (organic electroluminescent device).

[0015]

[Problems to be Solved by the Invention]

In the above-mentioned organic EL device utilizing phosphorescence have accompanied with a problem of luminescent deterioration particularly in an energized state. The reason for luminescent deterioration has not been clarified as yet but may be attributable to such a phenomenon that the life of triplet exciton is generally longer than that of singlet exciton by at least three digits, so that molecule is placed in a higher-energy state for a long period to cause reaction with ambient substance, formation of exciplex or excimer, change in minute

molecular structure, structural change of ambient substance, etc.

[0016]

Anyway, the (electro)phosphorescence device is expected to provide a higher luminescence efficiency but on the other hand, the device is problematic in terms of the (luminescent) deterioration in energized state.

[0017]

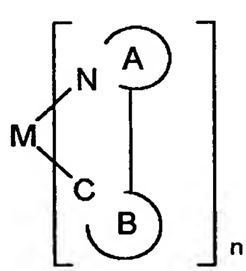
Accordingly, an object of the present invention is to provide a luminescence device capable of providing a high-efficiency luminescent, keeping a high brightness (or luminance) for a long period, and less deterioration (in luminescence) in energized state.

[0018]

[Means for Solving the Problems]

That is, the present invention provides a luminescence device characterized in that an organic compound layer comprising a metal coordination compound represented by the following formula (1):

[0019]



[0020]

{wherein M denotes Ir, Rh or Pd; n is 2 or 3; A and B are a nitrogen atom and a carbon atom, respectively, connected to a metal atom M; A and B are cyclic groups containing a nitrogen atom and a carbon atom, respectively, and capable of having a substituent, A and B being connected to each other via a covalent bond, [said substituent being a halogen atom; nitro group; a trialkylsilyl group (of which the alkyl group is a linear or branched alkyl group having 1 - 8 carbon atoms); or a linear or branched alkyl group having 1 - 20 carbon atoms (of which the alkyl group includes one or at least two non-neighboring methylene groups which can be replaced with -O-, -S-, -CO-, -CO-O-, -O-CO-, -CH=CH- or -C≡C- and includes a hydrogen atom which can be replaced with a fluorine atom] with the proviso that a sum of nitrogen atom and sulfur atom present in ring structures of A and B is at least 2}.

[0021]

In the luminescence device of the present invention, the metal coordination compound of formula (1) may preferably have a molecular structure free from a portion substantially causing intramolecular rotation and may preferably exhibit a peak (maximum) emission wavelength of at least 550 nm; the metal coordination compound of formula (1) may preferably

contain a ligand having a dipole moment of at most 7 debye, more preferably at most 4 debye, and may preferably exhibit a peak emission wavelength of at least 550 nm.

[0022]

Further, the present invention provides another luminescence device, including an organic compound layer comprising a metal coordination compound, wherein the metal coordination compound has a molecular structure free from a portion substantially causing intramolecular rotation and exhibits a peak emission wavelength of at least 550 nm.

[0023]

Further, the present invention provides another luminescence device, including an organic compound layer comprising a metal coordination compound, wherein the metal coordination compound contains a ligand having a dipole moment of at most 7 debye, preferably at most 4 days, and exhibits a peak emission wavelength of at least 550 nm.

[0024]

The luminescence device described above may preferably include two electrodes oppositely disposed to sandwich the organic compound layer to constitute an electroluminescence device, wherein a voltage is applied between the pair of electrodes to produce

luminescence.

[0025]

Further, the present invention provides a display apparatus, comprising: a luminescence device described above and a portion for driving the luminescence device.

[0026]

[Embodiments for Practicing the Invention]

In the case where a luminescence layer is formed of a carrier transporting host material and a phosphorescent guest material, a process of producing phosphorescence may generally involve the following steps:

(1) transport of electron and hole within a luminescence layer,

(2) formation of exciton of the host material,

(3) transmission of excited energy between host material molecules,

(4) transmission of excited energy from the host material molecule to the guest material molecule,

(5) formation of triplet exciton of the guest material, and

(6) production of phosphorescence during transition from the triplet excited state to the ground state of the guest material.

[0027]

In the respective steps, desired energy

transmission and luminescence may generally be caused based on various quenching and competition.

[0028]

In order to improve a luminescence efficiency of the EL device, it is needles to say that a luminescence center material per se provides a higher yield of luminescence quantum. However, an efficient energy transfer between host material molecules or between host material molecule and guest material molecule is also an important factor. Further, the reason for the above-described luminescent deterioration in energized state is not clarified as yet but may presumably relate to the luminescent center material per se or an environmental change thereof by its ambient molecular structure.

[0029]

For this reason, the present inventors have extensively investigated an effect of use of the metal coordination compound of formula (1) as the luminescent center material and have found that the compound produces a high-efficiency luminescence and retains a high brightness (luminance) for a long period.

[0030]

The metal coordination compound used in the present invention produces phosphorescence (luminescence) and is assumed to have a lowest excited

state which is an MLCT* (metal-to-ligand charge transfer) state as a triplet (excited state). The phosphorescence is produced during the transition from these states to the ground state.

[0031]

The luminescent material of the present invention has been found to provide a higher phosphorescence yield of 0.15 - 0.9 and a shorter phosphorescence life of 1 - 30 μsec.

The shorter phosphorescence life is a condition for providing a resultant EL device with a higher luminescence efficiency. More specifically, if the phosphorescence life is longer, molecules placed in their triplet excited state which is a waiting state for phosphorescence are increased to result in a problem of lowering the resultant luminescence efficiency particularly at a higher current density. The material of the present invention is a suitable luminescent material for an EL device with a higher phosphorescence yield and a shorter phosphorescence life. Further, due to realization of the shorter phosphorescence life leading to a shorter time period wherein the molecules stay in the triplet excited state, i.e. a higher energy state, so that the resultant EL device with improved durability and less deterioration in device characteristic.

[0032]

Actually, even in energization test, when the luminescent material of the present invention was used, it exhibited high stability.

[0033]

Further, in the ligand, according to the present invention, a plurality of nitrogen atom(s) and/or sulfur atom(s) in total in two ring structures, so that it becomes possible to decrease an energy gap, thus allowing a long-(emission) wavelength luminescence. This may be understood as follows.

In the case where HOMO (highest occupied molecular orbital) and LUMO (lowest unoccupied molecular orbital) of electron orbital for the metal and those (HOMO and LUMO) of electron orbital for the ligands are separately considered for convenience, energies of HOMO and LUMO for the ligands are lowered by the presence of nitrogen atom(s) and/or sulfur atom(s) to decrease an energy gap between HOMO for the metal and LUMO for the ligands, thus allowing luminescence from the $MLCT^*$ state being the lowest excited state at a longer (emission) wavelength. Accordingly, the metal coordination compound of the present invention is suitable luminescent material for luminescence at longer wavelength (orange to red).

[0034]

Further, in the case where a substituent is introduced into the ligand(s) as described above, an

intermolecular interaction is controlled when a device is prepared, thus reducing an occurrence of thermal deactivation of excited molecules and improving film uniformity, electrical properties and device stability.

[0035]

Further, the luminescent material of the present invention contains a plurality of nitrogen atom(s) and/or sulfur atom(s) in the ligands, so that electron clouds may presumably be considerably protruded from the ligands to promote interaction with other molecules via these atoms, thus improving energy transfer efficiency. As a result, it becomes possible to realize a luminescent material, for longer wavelength (orange to red) with high efficiency and stability, which has not been conventionally provided.

[0036]

Further, the metal coordination compound of the present invention is a coordination compound such that a plurality of identical ligands are coordinated. For example, yet-unreacted acetylacetonate ligand of iridium (III) acetylacetonate as a synthesis reaction initial compound used in Example 17 appearing hereinafter remains in the resultant complex to provide a compound, having different ligands which are coordinated, as a reaction product, in some cases. In the case of this compound, the compound has a poor

thermal stability and can cause thermal decomposition when an EL device is prepared through vacuum (vapor) deposition wherein the metal coordination compound is sublimated by resistance heating. Indeed the coordination compound containing acetylacetonate ligand has a lower initiation temperature for thermal decomposition. Further, due to the poor thermal stability, the life of the resultant (luminescence) device using such a compound becomes shorter. Further, different ligands are present in the device, so that electron levels (for the different ligands) are different from each other, thus adversely affecting electron transport performance.

[0037]

In the case of phosphorescent (luminescent) material, luminescent characteristics are largely depend on its molecular environment. In he case of the fluorescent material, essential properties of the luminescent material are studied based on photoluminescence. For this reason, results of photoluminescence of the phosphorescent material do not reflect luminescent characteristics of the resultant EL device in many cases since the luminescent characteristics in the case of the phosphorescent material depend on a magnitude of polarity of ambient host material molecules, ambient temperature, state of solid or liquid, etc.

Accordingly, it is generally difficult to expect the resultant EL characteristics (for the phosphorescent material) by simply removing a part of characteristics from photoluminescence results.

[0038]

On the other hand, as a result of our investigation on various phosphorescence metal coordination compounds represented by the luminescence material of the present invention, we have found that a high-efficiency luminescence can be achieved in the case where a metal coordination compound has a ligand having no portion causing intramolecular rotation and exhibits a peak (maximum) emission wavelength of at least 550 nm.

[0039]

The intramolecular rotation is a phenomenon such that atomic groups disposed opposite to each other via a single bond (as a rotation axis) in molecule are rotated relative to each other. The intermolecular rotation of ligand will be explained with reference to formula (1). The cyclic group A containing nitrogen atom and the cyclic group B containing carbon atom are bonded to each other via covalent bond, so that the covalent bond can function as an intramolecular rotation axis for the ligand alone. However, in the metal coordination compound, the nitrogen atom of A and the carbon atom of B are

bonded to the metal atom M, thus suppressing such an intermolecular rotation. Accordingly, the intramolecular rotation of ligand referred to herein means an intramolecular rotation on a single bond between a ring structure A and a substituent present thereat or on a single bond between a ring structure B and a substituent present thereat. A high-efficiency luminescence may be attributable to suppression of intramolecular rotation of ligand leading to a decrease in deactivation path within molecule caused by the intramolecular rotation at the time of luminescence of metal coordination compound.

Further, as a result of our investigation on ligands of various phosphorescence metal coordination compounds regarding a relationship between a luminescence efficiency and a dipole moment calculated based on a semi-empirical molecular orbital method (AM1), we have found that a high-efficiency luminescence is achieved in the case where a metal coordination compound contains a ligand having a dipole moment of at most 7 debye, preferably at most 4 debye, and exhibits a peak emission wavelength of at least 550 nm.

[0040]

This may be attributable to suppression of localization of electric charges of ligands, thus suppressing intermolecular interactions, e.g., between

metal coordination compound molecules and between a guest material molecule and a host material molecule thereby to decrease the deactivation path between their molecules to improve a luminescence efficiency. Further, the magnitude of dipole moment of ligand is a factor for determining a lowest excited state affecting a degree of luminescence efficiency. The lowest excited state includes the MLCT* (metal-ligand charge transfer) state and ππ* state. In many cases, the MLCT* state provides a relatively high phosphorescence efficiency. This may be attributable to such a phenomenon that the metal as a heavy atom directly affects during luminescence, thus effectively causing spin-orbit interaction to increase a resultant phosphorescence efficiency. When a ligand has a larger dipole moment, an excitation from a site having a larger (electric) charge distribution within the ligand to a site having a smaller charge distribution becomes advantageous. That is, the ππ* excited state is stabilized to be disadvantageous to improvement in phosphorescence efficiency as described above.

[0041]

Further, as shown in Examples appearing hereinafter, the compound of the present invention has been clarified to exhibit excellent performance even in terms of stability in an energization durability test.

[0042]

As shown in Figure 1, the luminescence device of the present invention may preferably be an electroluminescent device wherein an organic layer comprising a metal coordination compound is sandwiched between oppositely disposed two electrodes between which are supplied with a voltage to produce luminescence.

[0043]

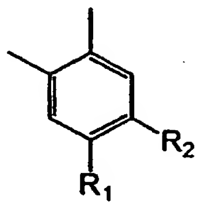
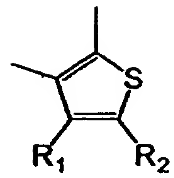
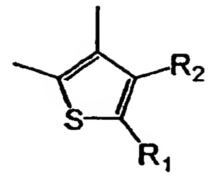
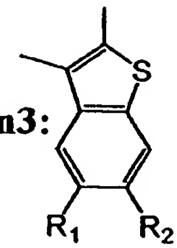
The high-efficiency luminescence device according to the present invention may be applicable to devices required to allow energy saving and high luminance. Applied examples thereof may include display apparatus and illumination apparatus, a light source for printers, and backlight (unit) for a liquid crystal display apparatus. As the display apparatus, it is possible to provide a flat panel display apparatus capable of exhibiting an excellent energy saving performance, a high visibility and a good lightweight property. With respect to the light source, it becomes possible to replace a laser light source of laser beam printer currently used widely with the luminescence device according to the present invention. An independently addressable device is arranged in an array and desired exposure of light to a photosensitive drum is effected to form an image. By using the device of the present invention, it

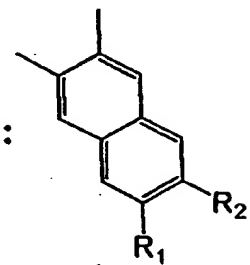
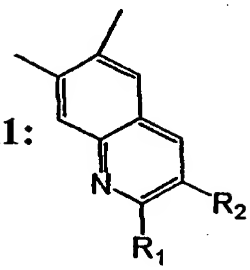
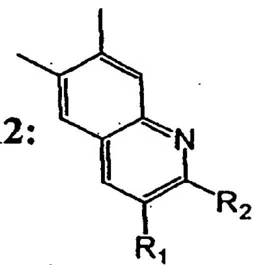
becomes possible to considerably reducing the volume (size) of image forming apparatus. With respect to the illumination apparatus and backlight (unit), an energy saving effect by the present invention can be expected.

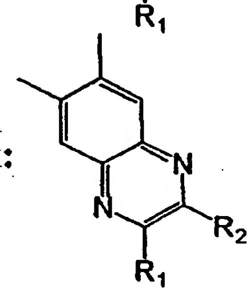
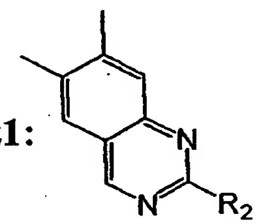
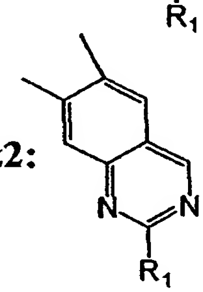
[0044]

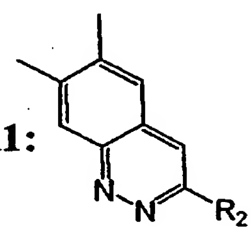
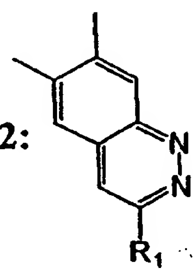
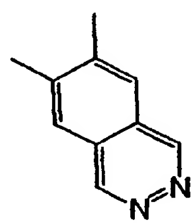
Hereinbelow, specific structural formulas of the metal coordination compound used in the present invention will be shown in Tables 1 - 9. However, these merely show representative examples and the present invention is not limited thereto. Ph to Pz used in Tables 1 - 9 represent structures shown below.

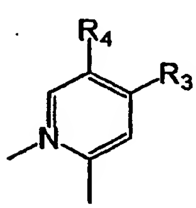
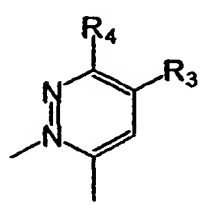
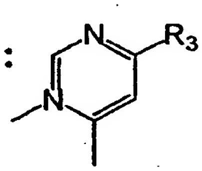
[0045]

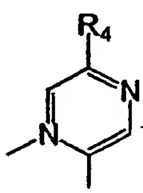
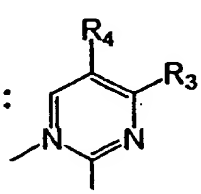
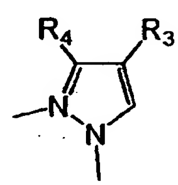
Ph:  **Tn1:**  **Tn2:**  **Tn3:** 

Np:  **Qn1:**  **Qn2:** 

Qx:  **Qz1:**  **Qz2:** 

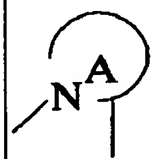
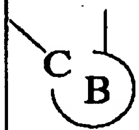
Cn1:  **Cn2:**  **Pz:** 

Pr:  **Pd:**  **Py1:** 

Pa:  **Py2:**  **Pz:** 

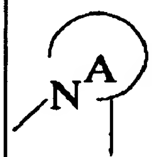
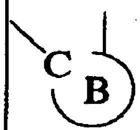
[0046]

[Table 1]

No	M	n			R1	R2	R3	R4
1	Ir	3	Pr	Tn1	H	H	H	H
2	Ir	3	Pr	Tn2	H	H	H	H
3	Ir	3	Pr	Tn3	H	H	H	H
4	Ir	3	Pr	Qn1	H	H	H	H
5	Ir	3	Pr	Qn2	H	H	H	H
6	Ir	3	Pr	Qx	H	H	H	H
7	Ir	3	Pr	Qz1	H	H	H	H
8	Ir	3	Pr	Qz2	H	H	H	H
9	Ir	3	Pr	Cn1	H	H	H	H
10	Ir	3	Pr	Cn2	H	H	H	H
11	Ir	3	Pr	Pz	H	H	H	H
12	Ir	3	Pd	Ph	H	H	H	H
13	Ir	3	Pd	Tn1	H	H	H	H
14	Ir	3	Pd	Tn2	H	H	H	H
15	Ir	3	Pd	Tn3	H	H	H	H
16	Ir	3	Pd	Np	H	H	H	H
17	Ir	3	Pd	Qn1	H	H	H	H
18	Ir	3	Pd	Qn2	H	H	H	H
19	Ir	3	Pd	Qx	H	H	H	H
20	Ir	3	Pd	Qz1	H	H	H	H
21	Ir	3	Pd	Qz2	H	H	H	H
22	Ir	3	Pd	Cn1	H	H	H	H
23	Ir	3	Pd	Cn2	H	H	H	H
24	Ir	3	Pd	Pz	H	H	H	H
25	Ir	3	Py1	Ph	H	H	H	H

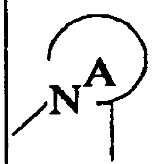
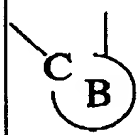
[0047]

[Table 2]

No	M	n			R1	R2	R3	R4
26	Ir	3	Py1	Tn1	H	H	H	H
27	Ir	3	Py1	Tn2	H	H	H	H
28	Ir	3	Py1	Tn3	H	H	H	H
29	Ir	3	Py1	Np	H	H	H	H
30	Ir	3	Py1	Qn1	H	H	H	H
31	Ir	3	Py1	Qn2	H	H	H	H
32	Ir	3	Pa	Ph	H	H	H	H
33	Ir	3	Pa	Tn1	H	H	H	H
34	Ir	3	Pa	Tn2	H	H	H	H
35	Ir	3	Pa	Np	H	H	H	H
36	Ir	3	Pa	Qn1	H	H	H	H
37	Ir	3	Pa	Qn2	H	H	H	H
38	Ir	3	Py2	Ph	H	H	H	H
39	Ir	3	Py2	Tn1	H	H	H	H
40	Ir	3	Py2	Tn2	H	H	H	H
41	Ir	3	Py2	Tn3	H	H	H	H
42	Ir	3	Py2	Np	H	H	H	H
43	Ir	3	Py2	Qn1	H	H	H	H
44	Ir	3	Py2	Qn2	H	H	H	H
45	Ir	3	Pz	Ph	H	H	H	H
46	Ir	3	Pz	Tn1	H	H	H	H
47	Ir	3	Pz	Tn2	H	H	H	H
48	Ir	3	Pz	Np	H	H	H	H
49	Ir	3	Pz	Qn1	H	H	H	H
50	Ir	3	Pz	Qn2	H	H	H	H

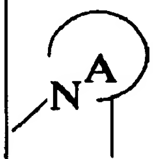
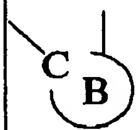
[0048]

[Table 3]

No	M	n			R1	R2	R3	R4
51	Rh	3	Pr	Tn1	H	H	H	H
52	Rh	3	Pr	Tn2	H	H	H	H
53	Rh	3	Pr	Tn3	H	H	H	H
54	Rh	3	Pr	Qn1	H	H	H	H
55	Rh	3	Pr	Qn2	H	H	H	H
56	Rh	3	Pr	Qx	H	H	H	H
57	Rh	3	Pr	Qz1	H	H	H	H
58	Rh	3	Pr	Qz2	H	H	H	H
59	Rh	3	Pr	Cn1	H	H	H	H
60	Rh	3	Pr	Cn2	H	H	H	H
61	Rh	3	Pr	Pz	H	H	H	H
62	Rh	3	Pd	Ph	H	H	H	H
63	Rh	3	Pd	Tn1	H	H	H	H
64	Rh	3	Pd	Tn2	H	H	H	H
65	Rh	3	Pd	Tn3	H	H	H	H
66	Rh	3	Pd	Np	H	H	H	H
67	Rh	3	Pd	Qn1	H	H	H	H
68	Rh	3	Pd	Qn2	H	H	H	H
69	Rh	3	Pd	Qx	H	H	H	H
70	Rh	3	Pd	Qz1	H	H	H	H
71	Rh	3	Pd	Qz2	H	H	H	H
72	Rh	3	Pd	Cn1	H	H	H	H
73	Rh	3	Pd	Cn2	H	H	H	H
74	Rh	3	Pd	Pz	H	H	H	H
75	Rh	3	Py1	Ph	H	H	H	H

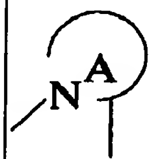
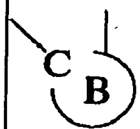
[0049]

[Table 4]

No	M	n			R1	R2	R3	R4
76	Rh	3	Py1	Tn1	H	H	H	H
77	Rh	3	Py1	Tn2	H	H	H	H
78	Rh	3	Py1	Tn3	H	H	H	H
79	Rh	3	Py1	Np	H	H	H	H
80	Rh	3	Py1	Qn1	H	H	H	H
81	Rh	3	Py1	Qn2	H	H	H	H
82	Rh	3	Pa	Ph	H	H	H	H
83	Rh	3	Pa	Tn1	H	H	H	H
84	Rh	3	Pa	Tn2	H	H	H	H
85	Rh	3	Pa	Np	H	H	H	H
86	Rh	3	Pa	Qn1	H	H	H	H
87	Rh	3	Pa	Qn2	H	H	H	H
88	Rh	3	Py2	Ph	H	H	H	H
89	Rh	3	Py2	Tn1	H	H	H	H
90	Rh	3	Py2	Tn2	H	H	H	H
91	Rh	3	Py2	Tn3	H	H	H	H
92	Rh	3	Py2	Np	H	H	H	H
93	Rh	3	Py2	Qn1	H	H	H	H
94	Rh	3	Py2	Qn2	H	H	H	H
95	Rh	3	Pz	Ph	H	H	H	H
96	Rh	3	Pz	Tn1	H	H	H	H
97	Rh	3	Pz	Tn2	H	H	H	H
98	Rh	3	Pz	Np	H	H	H	H
99	Rh	3	Pz	Qn1	H	H	H	H
100	Rh	3	Pz	Qn2	H	H	H	H

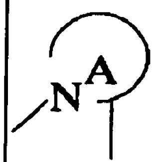
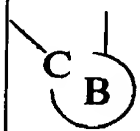
[0050]

[Table 5]

No	M	n			R1	R2	R3	R4
101	Pd	2	Pr	Tn1	H	H	H	H
102	Pd	2	Pr	Tn2	H	H	H	H
103	Pd	2	Pr	Tn3	H	H	H	H
104	Pd	2	Pr	Qn1	H	H	H	H
105	Pd	2	Pr	Qn2	H	H	H	H
106	Pd	2	Pr	Qx	H	H	H	H
107	Pd	2	Pr	Qz1	H	H	H	H
108	Pd	2	Pr	Qz2	H	H	H	H
109	Pd	2	Pr	Cn1	H	H	H	H
110	Pd	2	Pr	Cn2	H	H	H	H
111	Pd	2	Pr	Pz	H	H	H	H
112	Pd	2	Pd	Ph	H	H	H	H
113	Pd	2	Pd	Tn1	H	H	H	H
114	Pd	2	Pd	Tn2	H	H	H	H
115	Pd	2	Pd	Tn3	H	H	H	H
116	Pd	2	Pd	Np	H	H	H	H
117	Pd	2	Pd	Qn1	H	H	H	H
118	Pd	2	Pd	Qn2	H	H	H	H
119	Pd	2	Pd	Qx	H	H	H	H
120	Pd	2	Pd	Qz1	H	H	H	H
121	Pd	2	Pd	Qz2	H	H	H	H
122	Pd	2	Pd	Cn1	H	H	H	H
123	Pd	2	Pd	Cn2	H	H	H	H
124	Pd	2	Pd	Pz	H	H	H	H
125	Pd	2	Py1	Ph	H	H	H	H

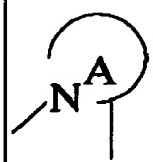
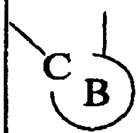
[0051]

[Table 6]

No	M	n			R1	R2	R3	R4
126	Pd	2	Py1	Tn1	H	H	H	H
127	Pd	2	Py1	Tn2	H	H	H	H
128	Pd	2	Py1	Tn3	H	H	H	H
129	Pd	2	Py1	Np	H	H	H	H
130	Pd	2	Py1	Qn1	H	H	H	H
131	Pd	2	Py1	Qn2	H	H	H	H
132	Pd	2	Pa	Ph	H	H	H	H
133	Pd	2	Pa	Tn1	H	H	H	H
134	Pd	2	Pa	Tn2	H	H	H	H
135	Pd	2	Pa	Np	H	H	H	H
136	Pd	2	Pa	Qn1	H	H	H	H
137	Pd	2	Pa	Qn2	H	H	H	H
138	Pd	2	Py2	Ph	H	H	H	H
139	Pd	2	Py2	Tn1	H	H	H	H
140	Pd	2	Py2	Tn2	H	H	H	H
141	Pd	2	Py2	Tn3	H	H	H	H
142	Pd	2	Py2	Np	H	H	H	H
143	Pd	2	Py2	Qn1	H	H	H	H
144	Pd	2	Py2	Qn2	H	H	H	H
145	Pd	2	Pz	Ph	H	H	H	H
146	Pd	2	Pz	Tn1	H	H	H	H
147	Pd	2	Pz	Tn2	H	H	H	H
148	Pd	2	Pz	Np	H	H	H	H
149	Pd	2	Pz	Qn1	H	H	H	H
150	Pd	2	Pz	Qn2	H	H	H	H

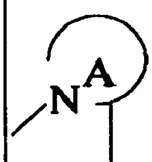
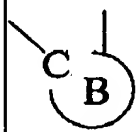
[0052]

[Table 7]

No	M	n			R1	R2	R3	R4
151	Ir	3	Pr	Tn1	H	$Si(CH_3)_3$	H	H
152	Ir	3	Pr	Tn1	H	SiC_2H_5	H	H
153	Ir	3	Pr	Tn1	H	OCH_3	H	H
154	Ir	3	Pr	Tn1	H	H	H	F
155	Ir	3	Pr	Tn1	H	H	F	F
156	Ir	3	Pr	Tn1	H	H	H	C_5H_{11}
157	Ir	3	Pr	Tn1	H	H	H	OCH_3
158	Ir	3	Pr	Tn1	H	H	OC_4H_9	H
159	Ir	3	Pr	Tn1	H	H	H	OCF_3
160	Ir	3	Pr	Tn1	H	H	H	SC_2H_5
161	Ir	3	Pr	Tn1	H	H	H	COC_2H_5
162	Ir	3	Pr	Tn1	H	H	H	$COOC_2H_5$
163	Ir	3	Pr	Tn1	H	H	H	$OCOC_2H_5$
164	Ir	3	Py1	Np	H	F	H	H
165	Ir	3	Pa	Np	H	OCH_3	H	H
166	Ir	3	Pz	Ph	H	H	H	CH_3
167	Ir	3	Pd	Ph	H	NO_2	H	H
168	Ir	3	Py1	Ph	H	NO_2	H	H
169	Ir	3	Pa	Ph	H	NO_2	H	H
170	Ir	3	Py2	Ph	H	F	H	H
171	Ir	3	Pz	Ph	H	F	H	H
172	Ir	3	Pz	Ph	H	$CH_2CH=CHCH_2CH_3$	H	H
173	Ir	3	Pz	Ph	H	$CH_2C\equiv CCH_2CH_3$	H	H
174	Rh	3	Pr	Tn1	H	$Si(CH_3)_3$	H	H
175	Rh	3	Pr	Tn1	H	OCH_3	H	H

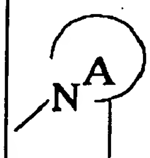
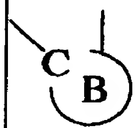
[0053]

[Table 8]

No	M	n			R1	R2	R3	R4
176	Rh	3	Pr	Tn1	H	H	H	F
177	Rh	3	Pr	Tn1	H	H	F	F
178	Rh	3	Pr	Tn1	H	H	H	C_5H_{11}
179	Rh	3	Pr	Tn1	H	H	H	OCH_3
180	Rh	3	Pr	Tn1	H	H	OC_4H_9	H
181	Rh	3	Pr	Tn1	H	H	H	OCF_3
182	Rh	3	Pr	Tn1	H	H	H	SC_2H_5
183	Rh	3	Pr	Tn1	H	H	H	COC_2H_5
184	Rh	3	Py1	Np	H	F	H	H
185	Rh	3	Pa	Np	H	OCH_3	H	H
186	Rh	3	Pz	Ph	H	H	H	CH_3
187	Rh	3	Pz	Tn1	H	F	H	H
188	Pd	2	Pr	Tn1	H	$Si(CH_3)_3$	H	H
189	Pd	2	Pr	Tn1	H	OCH_3	H	H
190	Pd	2	Pr	Tn1	H	H	H	F
191	Pd	2	Pr	Tn1	H	H	F	F
192	Pd	2	Pr	Tn1	H	H	H	C_5H_{11}
193	Pd	2	Pr	Tn1	H	H	H	OCH_3
194	Pd	2	Pr	Tn1	H	H	OC_4H_9	H
195	Pd	2	Pr	Tn1	H	H	H	OCF_3
196	Pd	2	Pr	Tn1	H	H	H	SC_2H_5
197	Pd	2	Pr	Tn1	H	H	H	COC_2H_5
198	Pd	2	Py1	Np	H	F	H	H
199	Pd	2	Pa	Np	H	OCH_3	H	H
200	Pd	2	Pz	Ph	H	H	H	CH_3
201	Pd	2	Pz	Tn1	H	F	H	H

[0054]

[Table 9]

No	M	n			R1	R2	R3	R4
202	Ir	3	Pr	Tn1	H	H	H	CF_3
203	Ir	3	Pr	Tn1	H	CH_3	H	H
204	Ir	3	Pr	Tn1	H	H	CF_3	H
205	Ir	3	Pr	Tn3	H	H	H	CF_3

[0055]

[Examples]

(Examples 1 - 10 and Comparative Example 1)

A common portion of device preparation steps employed in the present invention will be described.

[0056]

As a device structure, a device shown in Figure 1(b) having three organic layers was used. On a glass substrate (transparent substrate 15), a 100 nm-thick film (transparent electrode 14) of ITO (indium tin oxide) was formed, followed by patterning to have an opposing electrode area of 3 mm^2.

On the ITO-formed substrate, organic layers and metal electrode layers shown below were successively formed by vacuum (vapor) deposition using resistance heating in a vacuum chamber (10^{-4} Pa).

Organic layer 1 (hole transport layer 13) (40 nm): α-NPD

Organic layer 2 (luminescence layer 12) (30 nm): CBP:predetermined coordination compound (weight proportion: 5 wt. %)

Organic layer 3 (electron transport layer 16) (30 nm): Alq3

Metal electrode layer 1 (15 nm): Al-Li alloy (Li content = 1.8 wt. %)

Metal electrode layer 2 (100 nm): Al

[0057]

An electric field (voltage) was applied, so as to provide each device with the same current value, to each luminescence device having the ITO electrode (as an anode) and the Al electrode (as a cathode), to measure a change in luminance (brightness) with time. The current amount was set to 70 mA/cm^2 and the respective devices showed luminances in the range of 80 - 240 cd/m^2 at an initial stage required for decreasing.

[0058]

For measurement, each luminescence device was taken out of the vacuum chamber and was subjected to the measurement in an atmosphere of dry nitrogen gas stream so as to remove device deterioration factors, such as oxygen and moisture (water content).

[0059]

The results of continuous energization test of devices using the respective compound are shown in Table 10. As a comparative example, an experiment of the device using $Ir(ppy)_3$ as the luminescence material was conducted. The device (using $Ir(ppy)_3$ clearly provided shorter luminance half-life than the devices using the luminescent materials of the present invention, so that it becomes possible to provide a device having a high durability based on a stability of the materials of the present invention.

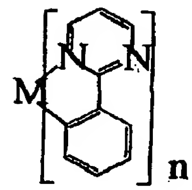
[0060]

[Table 10]

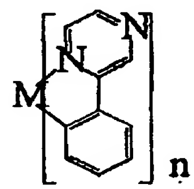
Ex. and Comp.Ex. No.	Luminescence material			Luminescence half-life
	Chem. formula No.	M (metal atom)	n	(hours)
Ex. 1	21	Ir	3	550
Ex. 2	22	Ir	3	900
Ex. 3	23	Ir	3	600
Ex. 4	24	Ir	3	650
Ex. 5	25	Ir	3	950
Ex. 6	26	Ir	3	800
Ex. 7	27	Ir	3	850
Ex. 8	28	Ir	3	600
Ex. 9	29	Ir	3	750
Ex. 10	30	Ir	3	900
Comp. Ex. 1		$Ir(ppy)_3$		350

[0061]

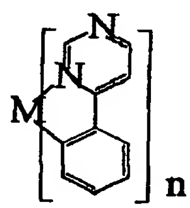
Chem. formula 21



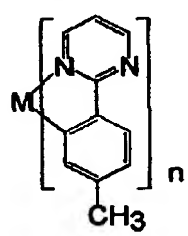
Chem. formula 22



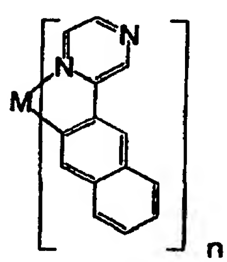
Chem. formula 23



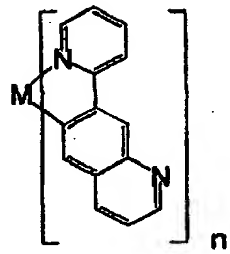
Chem. formula 24



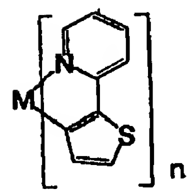
Chem. formula 25



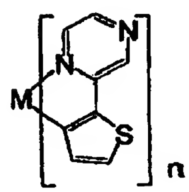
Chem. formula 26



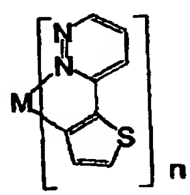
Chem. formula 27



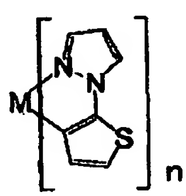
Chem. formula 28



Chem. formula 29



Chem. formula 30



[0062]

(Examples 11 - 16 and Comparative Example 2)

In these examples, as a device structure, a device having a four-layer structure of organic layers shown in Figure 1(c) was used. On a glass substrate (transparent substrate 15), a 100 nm-thick film (transparent electrode 14) of ITO (indium tin oxide) was formed, followed by patterning to have an (opposing) electrode area of 3 mm^2. On the ITO-formed substrate, organic layers and metal electrode layers shown below were successively formed by vacuum (vapor) deposition using resistance heating in a vacuum chamber (10^{-4} Pa).

Organic layer 1 (hole transport layer 13) (40 nm): α-NPD

Organic layer 2 (luminescence layer 12) (30 nm): mixture of CBP: predetermined metal coordination compound (weight proportion: 7 wt. %)

Organic layer 3 (exciton diffusion prevention layer 17) (10 nm): BCP

Organic layer 4 (electron transport layer 16) (30 nm): Alq3

Metal electrode layer 1 (15 nm): Al-Li alloy (Li content = 1.8 wt. %)

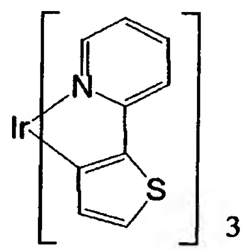
Metal electrode layer 2 (100 nm): Al

[0063]

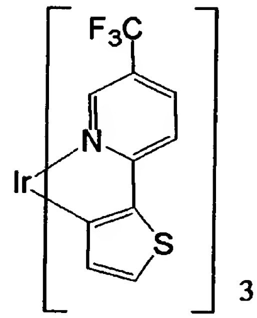
As the coordination compounds, compounds

represented by the following chemical formulas 41 - 46 were used.

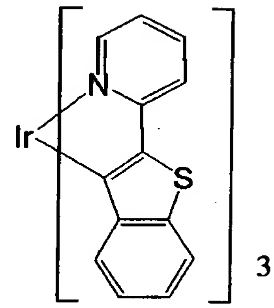
[0064]



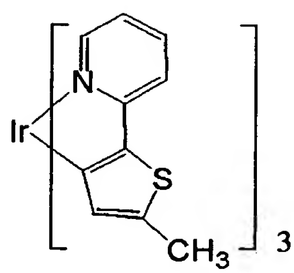
Chem. formula 41



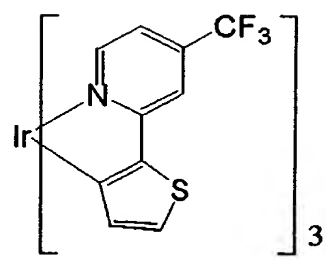
Chem. formula 42



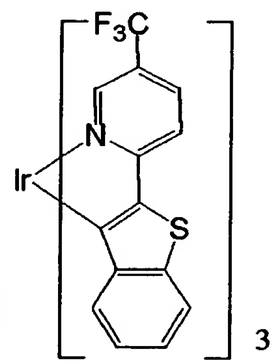
Chem. formula 43



Chem. formula 44



Chem. formula 45



Chem. formula 46

[0065]

Characteristics of the EL (luminescence) devices shown in Table 11 were measured by using a microammeter ("Model 4140B", mfd. by Hewlett-Packard Co.) for a current-voltage characteristic, using a spectrophotofluoro-meter ("Model SR1", mfd. by Topcon K.K.) for a luminescence spectrum, and using a

luminance meter ("Model BM7", mfd. by Topcon K.K.) for a luminescence brightness (luminance). The devices corresponding to the respective coordination compounds used in these examples showed a good rectification characteristic.

[0066]

Although $Ir(ppy)_3$ as the Comparative Example (2) showed a maximum of luminescence spectrum (peak emission wavelength) (λ_{PE}) = 510 nm, the EL devices using the metal coordination compounds used in these Examples were found to show longer peak emission wavelengths than $Ir(ppy)_3$ by ca. 4 - 110 nm. Further, with respect to the energy conversion efficiency and the luminescence efficiency, the present Examples provided smaller values than $Ir(ppy)_3$ but this is because a relative luminous efficiency becomes smaller with a longer wavelength. Accordingly, it is considered that the luminescence characteristic of the metal coordination compounds of these Examples is not essentially inferior to that of $Ir(ppy)_3$.

[0067]

As for the luminance half-lifes, an experiment was conducted in the same manner as in Examples 1 - 10. It is found that the luminescent materials according to Examples showed considerably longer luminance half-lifes than $Ir(ppy)_3$.

[0068]

[Table 11]

Ex.No	Coordination compound	Peak emission wavelength (nm)	Energy conversion efficiency (lm/W)	Luminescence efficiency (cd/A)	Current density (mA/cm^2 at 12V)	Luminance half-life (Hr)
Comp.Ex.2	$Ir(ppy)_3$	510	6.0	19.0	20	150
Ex.11	41	552	0.9	6.3	100	300
Ex.12	42	565	1.0	4.0	215	350
Ex.13	43	600	2.9	3.1	125	250
Ex.14	44	560	1.2	5.7	30	300
Ex.15	45	575	1.4	5.5	110	320
Ex.16	46	620	0.5	0.6	260	300

[0069]

When attention is directed to the substituent(s) causing intramolecular rotation, the compound of Chem. formula 41 which is free from a portion causing intramolecular rotation provided luminescence efficiency larger than the compounds of Chem. formulas 42, 44 and 45 which causing intramolecular rotation. Similarly, the compound of Chem. formula 43 which is free from a portion causing intramolecular rotation exhibited a luminance efficiency larger than that of the compound of Chem. formula 46 which is a portion causing intramolecular rotation. From these results, it was clarified that a metal coordination compound produced high-efficiency luminescence when its ligand had a ligand free from a portion causing intramolecular rotation and exhibited a peak emission wavelength of at least 550 nm.

[0070]

Hereinbelow, synthesis processes of the coordination compounds used in Examples 11 - 16 will be shown in Examples 17 - 22, respectively.

[0071]

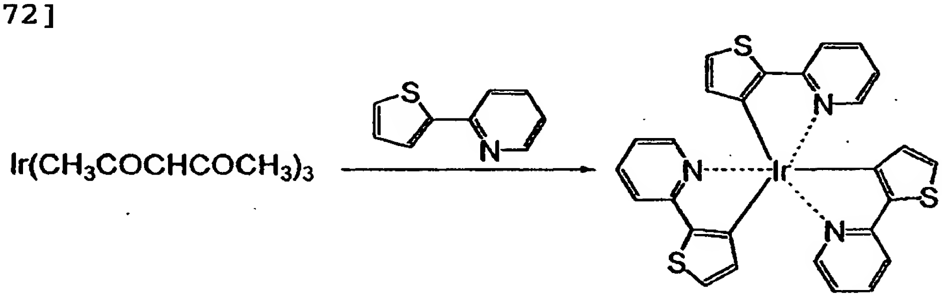
Example 17

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{o}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{o}C by

standing. To glycerol, 1.94 g (12.00 mM) of 2-(2-thienyl)pyridine (mfd. by Lancaster Co.) and 1.00 g (2.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 4 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) to obtain 0.38 g of Iridium (III) tris[2-(2-thienylphenyl)pyridine] (red powder) (Yield: 28.2 %).

[0072]



[0073]

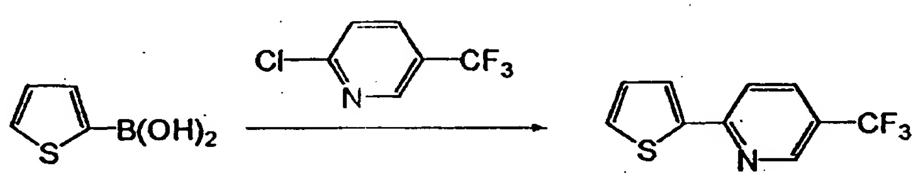
Example 18

In a 1 liter-three necked flask, 33.70 g (185.8 mM) of 2-chloro-5-trifluoromethylpyridine, 23.77 g (185.8 mM) of thiophene-2-bronic acid, 200 ml of toluene, 100 ml of ethanol and 200 ml of 2M-sodium carbonate aqueous solution were placed and stirred in

a nitrogen gas stream at room temperature. Under stirring, to the mixture, 6.66 g (5.76 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 5 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 2/1) and recrystallized from ethanol to obtain a pale yellow crystal. The crystal was purified by alumina column chromatography (eluent: toluene) and recrystallized from ethanol to obtain 20.3 g of 2-(5-trifluoromethylpyridine-2-yl)thiophene (Yield: 47.6 %).

[0074]



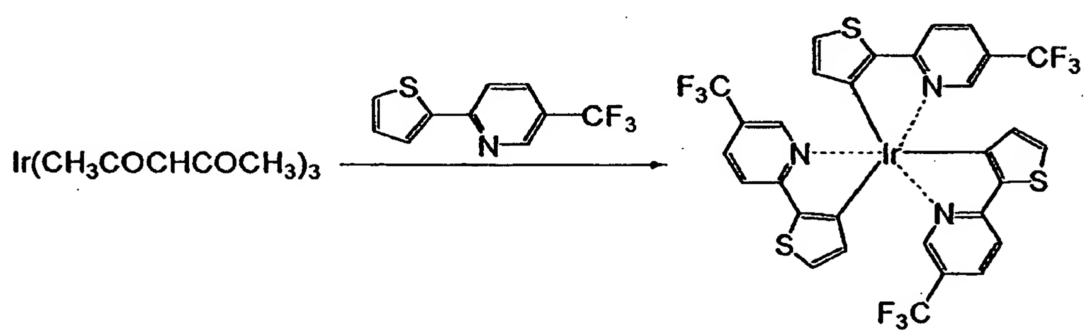
[0075]

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{O}C while supplying nitrogen gas therein in

the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 2.74 g (12.0 mM) of 2-(5-trifluoromethylpyridine-2-yl)thiophene and 1.00 g (2.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 4 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) to obtain 0.35 g of Iridium (III) tris[2-(5-trifluoromethylpyridine-2-yl)thiophene] (red powder) (Yield: 20.0 %).

[0076]



[0077]

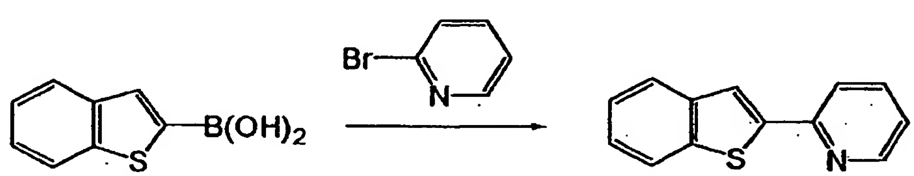
<u>Example 19</u>

In a 1 liter-three necked flask, 26.6 g (168.5 mM) of 2-bromopyridine, 30.0 g (168.5 mM) of benzo[b]thiophene-2-bronic acid, 170 ml of toluene, 85

ml of ethanol and 170 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 6.18 g (5.35 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 5.5 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 5/1) to obtain a colorless crystal. The crystal was purified by alumina column chromatography (eluent: toluene) and recrystallized from ethanol to obtain 12.6 g of 2-(pyridine-2-yl)benzo[b]thiophene (Yield: 35.4 %).

[0078]



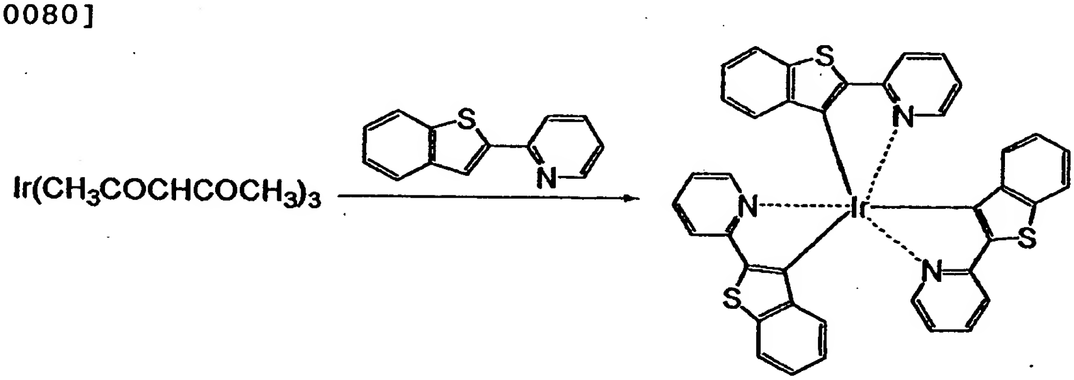
[0079]

In a 200 ml-four necked flask, 100 ml of

glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{o}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{o}C by standing. To glycerol, 2.52 g (12.0 mM) of 2-(pyridyl-2-yl)benzo[b]thiophene and 1.00 g (2.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration, washed with water, and dissolved in acetone followed by removal of the insoluble matter by filtration and distilling-off of acetone under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: chloroform) to obtain 0.38 g of Iridium (III) tris[2-(pyridine-2-yl)benzo[b]thiophene] (red powder) (Yield: 23.1 %).

[0080]



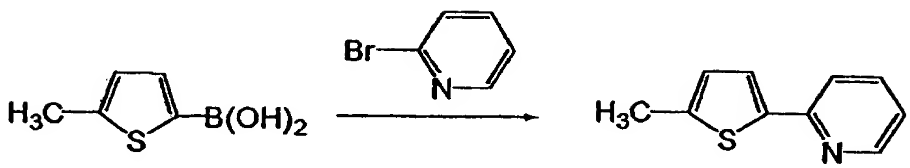
[0081]

Example 20

In a 1 liter-three necked flask, 27.0 g (171.2 mM) of 2-bromopyridine, 24.3 g (171.2 mM) of 5-methylthiophene-2-bronic acid, 180 ml of toluene, 90 ml of ethanol and 180 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 6.42 g (5.55 mM) of tetrakis (triphenyl-phosphine) palladium (0) was added, followed by heat-refluxing for 9 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene. The organic layer was washed with water until the system showed neutral, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: toluene/hexane = 5/1) to obtain a pale yellow crystal. The crystal was purified by alumina column chromatography (eluent: toluene) and successively recrystallized from ethanol and hexane to obtain 12.4 g of 2-(pyridine-2-yl)-5-methylthiophene (colorless crystal) (Yield: 41.3 %).

[0082]

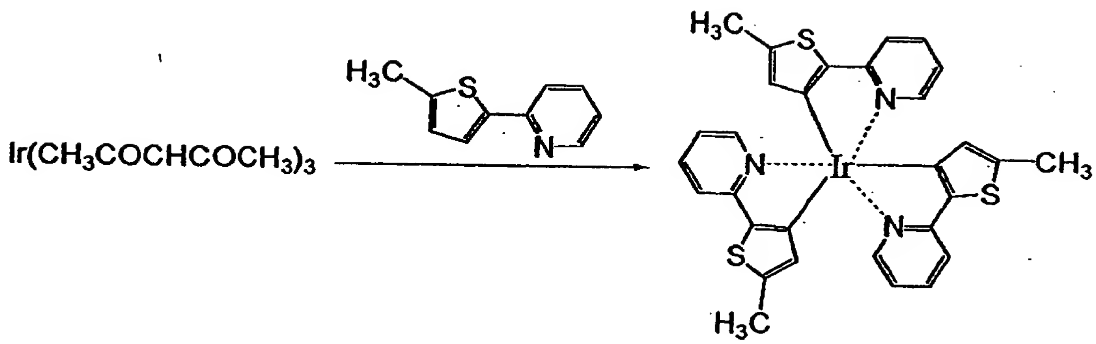


[0083]

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 °C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 2.10 g (12.0 mM) of 2-(pyridyl-2-yl)-5-methylthiophene and 1.00 g (2.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration, washed with water, and dissolved in acetone followed by removal of the insoluble matter by filtration and distilling-off of acetone under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: chloroform) to obtain 0.31 g of Iridium (III) tris[2-(pyridine-2-yl)-5-methylthiophene] (red powder) (Yield: 21.7 %).

[0084]



[0085]

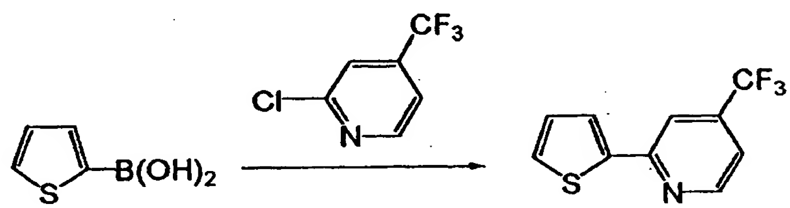
Example 21

In a 100 ml-three necked flask, 1.73 g (0.95 mM) of 2-chloro-4-trifluoromethylpyridine, 1.23 g (0.96 mM) of thiophene-2-bronic acid, 15 ml of toluene, 7.5 ml of ethanol and 15 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 0.34 g (0.30 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 4 hours and 10 minutes under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled, followed by extraction with cool water and toluene, followed by distilling off of the solvent under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: chloroform) to obtain a pale yellow crystal. The crystal was purified by alumina column chromatography (eluent: toluene) and

recrystallized from methanol to obtain 1.98 g of 2-(trifluoromethylpyridine-2-yl)thiophene (Yield: 91.2 %).

[0086]



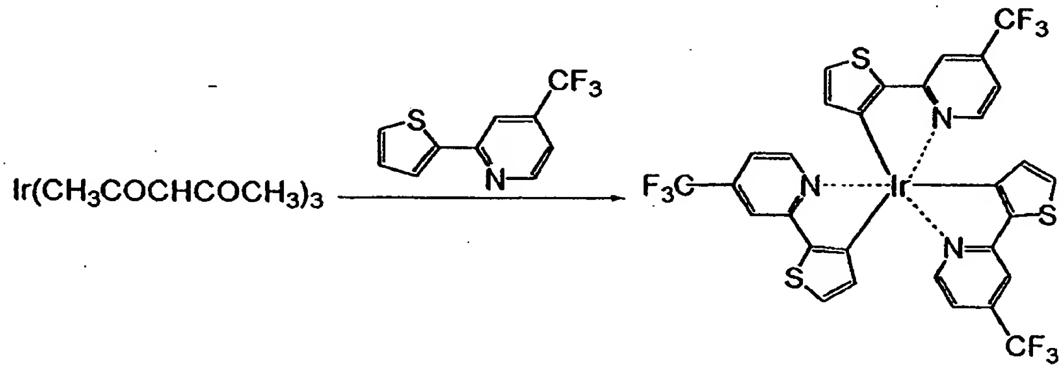
[0087]

In a 100 ml-four necked flask, 50 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 °C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 °C by standing. To glycerol, 1.20 g (5.2 mM) of 2-(4-trifluoromethylpyridine-2-yl)thiophene and 0.50 g (1.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 300 ml of 1N-HCl. The resultant precipitate was recovered by filtration and washed with water, followed by drying for 4 hours at 100 °C under reduced pressure and purification by silica gel column chromatography (eluent: chloroform) to obtain 0.15 g of Iridium (III) tris[2-(4-trifluoromethylpyridine-2-yl)thiophene] (red

powder) (Yield: 17.1 %).

[0088]



[0089]

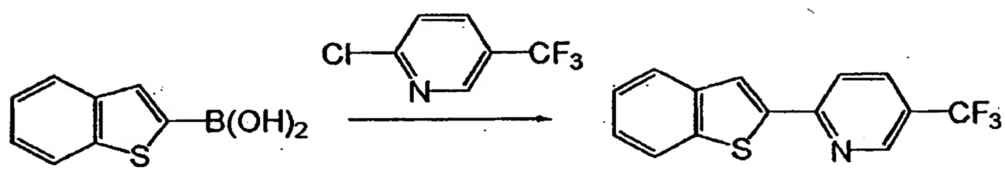
Example 22

In a 200 ml-four necked flask, 5.16 g (28.4 mM) of 2-chloro-5-trifluoromethylpyridine, 5.06 g (28.4 mM) of benzo[b]thiophene-2-bronic acid, 25 ml of toluene, 12.5 ml of ethanol and 25 ml of 2M-sodium carbonate aqueous solution were placed and stirred in a nitrogen gas stream at room temperature. Under stirring, to the mixture, 1.02 g (0.88 mM) of tetrakis (triphenylphosphine) palladium (0) was added, followed by heat-refluxing for 7.5 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled on an ice bath to precipitate a crystal. The crystal was recovered by filtration, followed by washing with water and further washing with methanol. The crystal was purified by alumina column chromatography (eluent: chloroform) and recrystallized

from chloroform to obtain 2.90 g of 2-(5-trifluoromethylpyridine-2-yl)benzo[b]thiophene (Yield: 36.5 %).

[0090]



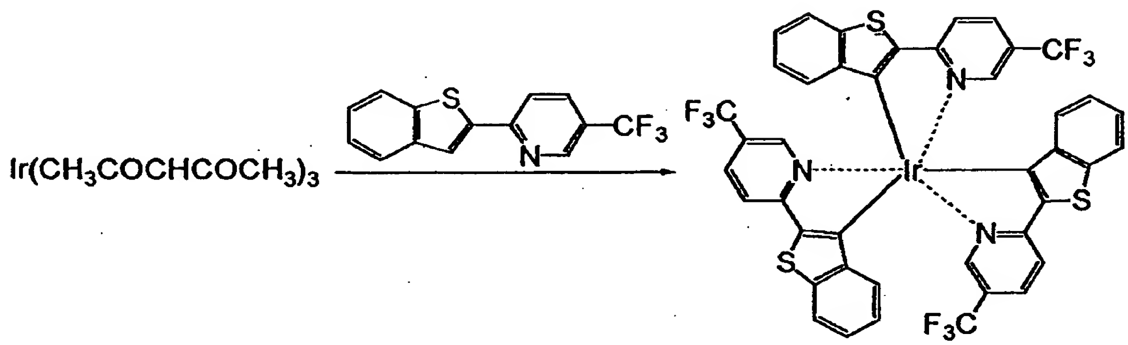
[0091]

In a 200 ml-four necked flask, 100 ml of glycerol was placed and heat-stirred for 2 hours at 130 - 140 ^{O}C while supplying nitrogen gas therein in the form of bubbles, followed by cooing to 100 ^{O}C by standing. To glycerol, 2.82 g (10.1 mM) of 2-(5-trifluoromethylpyridine-2-yl)benzo[b]thiophene and 1.00 g (2.0 mM) of Iridium (III) acetylacetonate were added, followed by heat-refluxing for 8 hours under stirring in nitrogen gas stream.

After the reaction, the reaction mixture was cooled to room temperature and poured into 600 ml of 1N-HCl. The resultant precipitate was recovered by filtration, washed with water, and dissolved in acetone followed by removal of the insoluble matter by filtration and distilling-off of acetone under reduced pressure to obtain a residue. The residue was purified by silica gel column chromatography (eluent: chloroform) to obtain 0.24 g of Iridium (III) tris[2-

(5-trifluoromethylpyridine-2-yl)benzo[b]thiophene] (red powder) (Yield: 11.4 %).

[0092]



[0093]

Example 23

With respect to ligands of the metal coordination compounds of Chem. formulas 41, 42, 43 and 46, dipole moments were calculated based on the semi-empirical molecular orbital method (AM1) as to the case where a conformation of respective ligands was such that respective two ring structures were present in an identical plane, thereby to investigate a relationship thereof with a luminescence efficiency. The results are shown in Table 12.

[0094]

[Table 12]

Metal coordination No.	Dipole-moment (debye)	EL device characteristics	
		λ_{PE} (nm)	Luminescence efficiency (cd/A)
41	1.8	552	6.3
42	3.7	565	4.0
43	1.3	600	3.1
44	8.6	620	0.6

[0095]

From this results, it has been found that a high-efficiency luminescence was realized in the case where the dipole moment was at most 7 debye, preferably at most 4 debye, and the peak emission wavelength (λ_{PE}) was at least 550 nm.

[0096]

[Effect of the Invention]

As described hereinabove, the metal coordination compound used in the present invention has a higher phosphorescence luminescence efficiency and a shorter phosphorescence life, thus being suitable for longer-waveform luminescence (orange to red) and being suitable as a luminescence material for an EL device.

[0097]

Further, the luminescence device using the

metal coordination compound according to the present invention is an excellent device which produces a high-efficiency luminescence, retains a high luminescence for a long period of time, and provides less deterioration in energized state.

[Brief Description of the Drawings]

Figure 1 is a schematic sectional view of a layer structure of a luminescence device of the present invention.

[Explanation of Symbols]

11: metal electrode

12: luminescence layer

13: hole transport layer

14: transparent electrode

15: transparent substrate

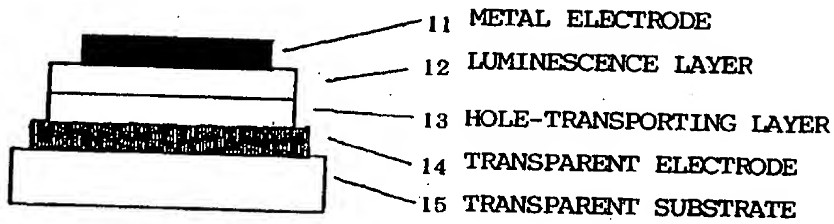
16: electron transport layer

17: exciton diffusion prevention layer

【DOCUMENT NAME】 DRAWING

FIG. 1

(a)



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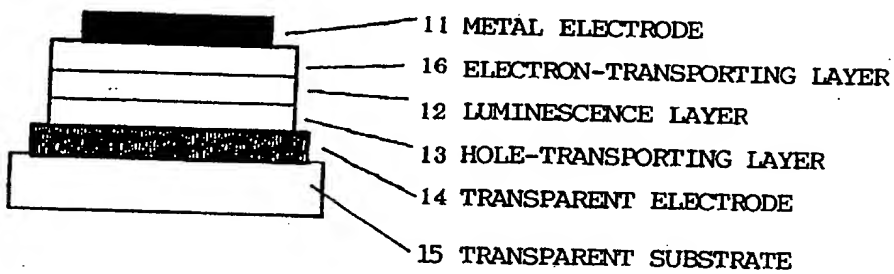

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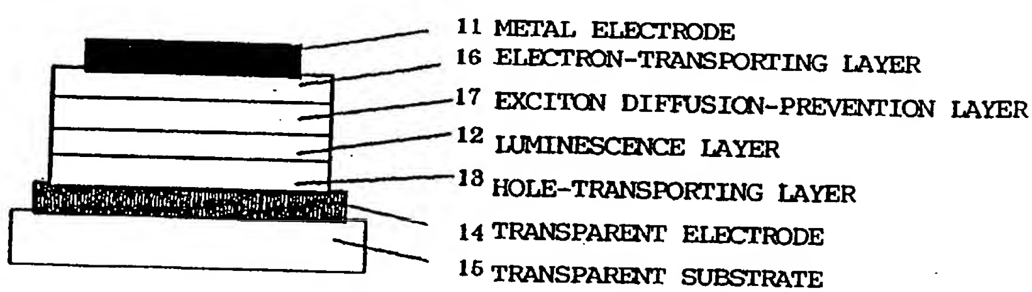


[Document]

Abstract

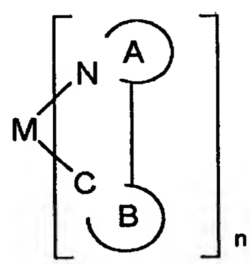
[Abstract]

[Problem]

To provide a luminescence device which produces a high-efficiency luminescence, maintains a high luminance for a long period of time, and provides less deterioration in energized state.

[Solution Means]

A luminescence device characterized by including an organic compound layer containing a metal coordination compound represented by the following formula (1):



(1)

[Selected Figure]

Figure 1